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Agent's File KT196F290.US
U.S. Patent Application No.08/957,187

DECLARATION

I, Dr. Wilfried Hatke, state that I am a resident of Am Waldeck 42a , 65779 Kelkheim/Ts, Federal Republic of Germany;
that I am a chemist having graduated at Philipps University Marburg, Marburg, Federal Republic of Germany;
that I am a co-inventor of U.S. Patent Application Serial No. 08/957,187 (hereinafter "the instant patent application") by DI Ekkehard Beer and myself for "Mono- or multilayer film"
that I have studied the contents of U.S. Patent No. 5,532,030 (hereinafter "the Hirose et al. patent") and that I am familiar with the subject matter thereof;
that I consider myself qualified by my knowledge in chemistry and especially in the fields of polymer chemistry and of polymer processing and by my 10 years experience in these fields;
that I have supervised and evaluated the following experiments and observations to wit:

Experimental:

In the experiments two copolymers were used which were obtained by polymerisation of ethylene and 2-norbornene in the presence of a metallocene catalyst. The copolymers were characterized by the following features:

Copolymer 1

- ethylene content of 45 mol% (determined via ^{13}C -NMR spectroscopy)
- glass transition temperature of 140°C (determined via differential scanning calorimetry (DSC) using a heating rate of 20°C/min and evaluating the midpoint of transition temperature)
- solution viscosity of 58 ml/g (determined in a 0.5 by weight % solution of the polymer in decaline at 135°C)
- molecular weight M_w of 42.000g/mol and M_n of 19.500g/mol (determined via gel permeation chromatography (GPC) using polyethylene standards and o-dichlorobenzene as an eluent; T=135°C)

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Copolymer 2

- ethylene content of 55 mol% (determined via ^{13}C -NMR spectroscopy)
- glass transition temperature of 80°C (determined via differential scanning calorimetry (DSC) using a heating rate of 20°C/min and evaluating the midpoint of transition temperature)
- solution viscosity of 80 ml/g (determined in a 0.5 by weight % solution of the polymer in decaline at 135°C)
- molecular weight M_w of 52.000g/mol and M_n of 24.500g/mol (determined via gel permeation chromatography (GPC) using polyethylene standards and o-dichlorobenzene as an eluent; $T=135^\circ\text{C}$)

Film preparation was performed by using conventional film processing equipment and applying different stretching conditions. Thus the polymer was transformed to a film by utilizing a single screw extruder equipped with a flat film die and a take up unit. The extruded films were subsequently stretched in a discontinuous stretcher (manufacturer: Brückner, Siegsdorf, Germany)

Results:

Stretching conditions

Example No.	polymer type	stretching factor	film thickness (prior to stretching) (μm)
1	1	0	80
2	1	3*3 ¹⁾	300
3	1	1.2*3.0 ¹⁾	100
4	1	1.2*3.0 ¹⁾	100
5	2	0	200
6	2	1.1	200
7	2	1.4	200
8	2	2.0	200
9	Blend	2.0	200
10	Blend	2.9	200
11	Blend2	3.5	200

¹⁾ biaxially stretched; Blend: Mixture of polymer 2 (75 weight-%) with polypropylene (melting point: 160°C, MFI: 9ml/10 min., 25 weight-%)

Film Properties

Example No.	modulus of elasticity (MPa)	tear strength (MPa)	elongation at break (%)	puncture resistance (N/mm)
1	2.350	60	2	60
2	3.350	92	45	550
3	2.500 ²⁾	55 ²⁾	2 ²⁾	120 ²⁾
4	3.600 ³⁾	95 ³⁾	30 ³⁾	120 ³⁾
5	2.050	65	3-4	200
6	2.500 ⁴⁾	55 ⁴⁾	2 ⁴⁾	210 ⁴⁾
7	3.600 ⁴⁾	95 ⁴⁾	45 ⁴⁾	245 ⁴⁾
8	3.600 ⁴⁾	95 ⁴⁾	45 ⁴⁾	290 ⁴⁾
9	2.350 ⁵⁾	110 ⁵⁾	70 ⁵⁾	300 ⁵⁾
10	2.450 ⁵⁾	115 ⁵⁾	90 ⁵⁾	n. determined
11	2.600 ⁵⁾	120 ⁵⁾	110 ⁵⁾	1.100 ⁵⁾
11a	2.050-2.100 ⁶⁾	40-45 ⁶⁾	3-4 ⁶⁾	n. determined

2) determined in direction of stretch factor 1.2

3) determined in direction of stretch factor 3.0

4) determined in direction of stretch factor 1.1 (Ex.6), 1.4 (Ex.7) and 2.0 (Ex. 8)

5) determined in direction of stretch factor 2.0 (Ex.9), 2.9 (Ex.10) and 3.5 (Ex. 11); p

6) determined in right angle direction to direction of stretching (films of Ex. 9, 10 and 11)

Conclusions:

From the comparison of Experiments 1 and 2-4 I conclude that stretching of a non-oriented film will increase the values of modulus of elasticity, tear strength, elongation at break and puncture resistance.

Comparison of Experiments 3 and 4 shows that for modulus of elasticity, tear strength and elongation at break the increase of said properties depends on the degree of stretching and that puncture resistance remains unaffected.

Comparison of experiment 4 with experiment 2 shows that a film stretched in both directions (experiment 2) has a significantly improved puncture resistance although the degree of stretching in each direction is similar. Nevertheless modulus of elasticity, tear strength and elongation at break of both films are essentially the same.

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demonstrate that modulus of elasticity, tear strength and elongation at significantly increasing in the direction of stretching for a second polym. ure resistance of these films is remaining at a low level.

Experiments 9-11 demonstrate that applying stretch ratios above 3 increase puncture resistance also for films stretched only in one direction.

In the Hirose et al. patent there is a general teaching to subject polyolefin multilayer laminate films to mono- or biaxial stretching (col. 34, l. 42-45). Such processing is known to those skilled in the art for improvement of mechanical properties of polymer films.

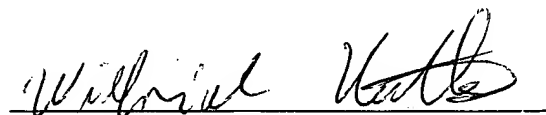
But from Hirose et al. patent it can not be concluded that for a desired value of puncture resistance the degree of stretching in combination with the direction(s) of stretching have to be carefully chosen as demonstrated in the instant specification.

Thus I conclude that the observed film properties are based on a surprising effect and that the instantly claimed subject matter can not be predicted from the general teaching of Hirose et al. patent.

Final Statement:

I declare that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements are made with the knowledge that wilful false statements and the like so made are punishable by fine of imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such wilful false statements may jeopardize the validity of the application or any patent issuing thereon.

Frankfurt am Main, this 08th day of May, 2001



Dr. Wilfried Hatke

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